Ultrafast Single and Multiple Electron Transfer from Colloidal Quantum Dots

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Charge transfer to and from quantum dots (QDs) is of intense interest because of its important roles in QD-based devices, such as solar cells and light emitting diodes. Recent reports of multiple exciton generation (MEG) by one absorbed photon in some QDs offer an exciting new approach to improve the efficiency of QD-based solar cells and to design novel multi-electron/hole photocatalysts. However, two main challenges remain. First, the efficiency of MEG process remains controversial and may need to be significantly improved for practical applications. Second, the utilization of the MEG process requires ultrafast exciton dissociation prior to the exciton-exciton annihilation process, which occurs on the 10's to 100's ps time scale. In this presentation we report a series of studies of exciton dissociation dynamics in quantum dots by electron transfer to adsorbed electron acceptors. We show that excitons in CdSe and PbS QDs can be dissociated on the a few picosecond timescale to various adsorbates. As a proof of principle, we demonstrated that multiple excitons (generated by absorbing multiple photons) per QD can be dissociated by electron transfer to adsorbed acceptors (*J. Am. Chem. Soc.*, 132, 4858-4864, [2010]). We will discuss the dependence of these rates on the size and the nature of the quantum dots and possible approaches to optimize the multiple exciton dissociation efficiency.